Models for the Angle-Dependent Optical Properties of Coated Glazing Materials

*M. Rubin, *R. Powles and **K. von Rottkay*

Windows and Daylighting Group
Building Technologies Department
Environmental Energy Technologies Division
Earnest Orlando Lawrence Berkeley National Laboratory
University of California
Berkeley, CA 94720

*School of Physics
University of Sydney
NSW 2006, Australia

**Physics Institute
Justus-Liebig-University Giessen
35392 Giessen, Germany

March 1998

Models for the Angle-Dependent Optical Properties of Coated Glazing Materials

M. Rubin

Lawrence Berkeley National Laboratory, University of California,
1 Cyclotron Road, MS2-300, Berkeley CA 94720, USA

R. Powles

School of Physics, University of Sydney, NSW 2006, Australia

K. von Rottkay

Physics Institute, Justus-Liebig-University Giessen, 35392 Giessen, Germany

Abstract

Optical transmittance and reflectance of window materials can be measured accurately at normal incidence using standard equipment. Sunlight often strikes at angles for which the transmittance and reflectance are significantly different from their values at normal incidence. A reliable procedure for extrapolating from normal properties to oblique properties is thus needed for accurate annual energy performance calculations and product comparisons. The structural models for the materials are greatly constrained by the limited amount of data that is usually available. For monolithic materials such as uncoated glass or plastic substrates it is possible to solve directly for the optical indices and then apply Fresnel's equation to obtain the oblique properties. For coated glass, the situation is more complex, but a numerical solution is often possible. First, detailed optical models were constructed and accurate angle-dependent data was generated for a wide selection of coated glazing materials. Then, a set of very simple thin-film models were chosen that would converge given a limited amount of data. At 60° incidence, the monolithic model was often accurate to within 2% but frequently deviated farther, up to 8%. The single-layer thin-film model fared little better. Highly constrained multilayer models often deviated less than 1 % although convergence became increasingly specific to similar coating types.

1. Introduction

Optical and radiative properties of glazing materials are primary inputs for determination of energy performance in buildings. Direct measurement of all relevant quantities for all products is not a practical option. Spectral transmittance and reflectance at normal incidence are the most easily and commonly measured properties. For uncoated glazing, it is also possible to determine angle dependence, even with this limited data (Rubin *et al.,* 1988*)*. Angle dependence of coated glazing requires a more complex numerical treatment

in order to obtain acceptable results. Furthermore, solutions to other problems, such as the synthesis of laminated structures, can be found within the same framework.

Direct calculations of angle-dependent optical properties could be performed in a straightforward and uniform way if the optical indices were known at every point within the system. Fresnel's equations and analogous equations for thin-film structures provide solutions for any plane-parallel window system. General application of this fundamental approach is usually not possible because of lack of complete and accurate knowledge of the coating. At the other extreme, the radiometric properties could be directly measured at each angle and polarization for any combination of layers and coatings. This approach is impractical, however, not only because few laboratories can currently measure angle-dependent properties, but also because of a ten-fold increase in measurement effort. Furthermore, oblique measurements and calculations do not yet fall within the scope of existing standards.

Although the fundamental optical parameters of coated window materials are not readily available, they can be extracted in principle from radiometric data at normal incidence. Some of the most complex coated glazing structures have been successfully analyzed using both ellipsometric and radiometric data. In general, however, the determination of the optical constants is a time-consuming and specialized operation. Furthermore, the numerical process is not fail-safe and will not automatically converge to a correct or satisfactory solution. It is possible, however, to arrive at approximate solutions that meet most of the required criteria without necessarily resulting in the most precise values for the optical indices themselves.

This paper begins by selecting a set of coating types representative of the broad range of available commercial products. An accurate dataset is first established, based on both experiment and theory, for the angle-dependent properties of these coatings. Structural and physical models of varying degrees of complexity are numerically fitted to spectral radiometric data limited to normal incidence. This process results in a set of effective optical indices and thickness that can be used to predict the properties at oblique angles. Finally, the properties predicted from normal incidence data are compared to the accurate data in an attempt to find a predictive scheme of general usefulness.

2. Experimental Methods

Transmittance and reflectance measurements are made over the full solar spectrum (250-2500 nm) at normal incidence using a Perkin-Elmer Lambda 19 spectrophotometer. The radiometric measurements and spectral averages were made in accordance with the standard practice of NFRC (1994), which is based in turn on the technical procedures of ASTM (1988). Round-robin tests among U.S. manufacturers produced repeatability better than 1% in transmittance by following the NFRC procedures. We hope to achieve the same level of accuracy in predicted angular properties. A variable-angle spectroscopic ellipsometer by J.A Woollam Co. provided supplemental data for unrestricted models only. The range of the ellipsometer covered the visible spectrum and part of the solar infrared (280-1700 nm).

Directional radiometric measurements were made at 670 nm using a feedback-stabilized laser diode with a short coherence length and a large-area silicon detector. The error for this system is typically 0.1%. Simplicity often translates to accuracy; the single light path, collimated laser beam and large-area uniform detector make systematic errors much easier to analyze and eliminate. Agreement has been demonstrated that is generally better than 1% between the spectroscopic goniometer at Uppsala University and our fixed-wavelength goniometer, although larger systematic discrepancies were noted for reflectance at high angles (Roos, 1997). In Section 3.2 it will be seen that these measurements are in excellent agreement with the predictions of our most detailed models.

3. Methods and Models

3.1 Representative Coating Types

One of our objectives is that the methods developed herein should produce acceptable accuracy for any existing glazing. Of course, a completely new category of coating may arise, in which case the method should prove adaptable. In the Window 4.1 database (Optical Data Library, 1998) there are currently about 750 product files including most if not all major coating categories. A rough estimate of the worldwide total number of basic products (not including permutations like laminates) might someday bring the total to 1500 files. Practically, we cannot process 1500 models, so we must look for far smaller number of categories. We also seek to determine how well each of the simplified models works when applied to various coating types. A small test set of glazing materials has been assembled, representative of commonly available coating types.

Roos (1997) broadly categorized the coating materials in common use: conducting oxides, noble metals, transition metals and semiconductors. He showed that the angular dependence of a given coating may not depend greatly on the particular material within a category. Pfrommer *et al.* (1995) demonstrated this insensitivity to material type for the specific case of noble metals. We do not share the view, mentioned in several papers, that the angular profile is weakly dependent on the thickness of absorbing films. In fact we find that film thickness can be one of the strongest factors, along with the specific coating configuration.

Montecchi and Polato (1995) created a useful test set of glazing materials with a range of optical characteristics. We found close analogues to most of the materials in that set (see Table 1) in our Window 4.1 Spectral Data Library. We previously modeled coatings similar to the pyrolytic tin oxide (von Rottkay and Rubin, 1996). The silver-based low-emittance coating included by Montecchi and Polato was supplemented by a double-silver spectrally selective coating, as well as a hybrid low-emittance coating with a thicker silver layer having reduced solar transmittance overall. Although the materials involved are the same in each of the three coatings, it was considered that the different thickness and coating structures might put each one in a different category of angle dependence.

We did not include the sol-gel "mixed oxide" coatings from the original set. The component oxides were not specified, and we were not aware of any coatings deposited by sol-gel techniques among the commercial glazing products. We have, however, successfully modeled experimental sol-gel coatings for electrochromic applications (Rubin

et al., 1996; von Rottkay *et al.,* 1995). Also, we were unable to discover pyrolytic (or other) coatings containing only "cobalt and iron oxides" or "chromium and iron oxides" (Montecchi *et al.*, 1994) in our database of products. Products consisting of pyrolytic Co-Fe-Cr oxides in roughly equal proportion were identified too late for inclusion in this paper, and they too have been previously modeled (Ruzakowski *et al.*, 1997; Ruzakowski *et al.,* 1996).

Table 1. Test Set from Window 4.1 Spectral Data Library.

Principal Optical Components	Window 4.1 Filename	T_{vis}	R_{vis}	T_{sol}	R_{sol}	E
Titanium Nitride (TiN)	B130.AFG	0.308	0.274	0.247	0.300	0.61
Stainless Steel Fe-Cr-Ni (SS)	P120.AFG	0.236	0.273	0.211	0.289	0.58
TiN/ Fe-Cr-Ni (TiN/SS)	S120.AFG	0.216	0.321	0.178	0.380	0.52
Thin Ag / dielectric (D/Ag/D)	E78_4.CIG	0.860	0.048	0.695	0.148	0.08
Thicker Ag/ dielectric (D/Ag+/D)	SUN45_4.CIG	0.494	0.135	0.387	0.175	0.10
Double Ag/dielectric (D/Ag/D/Ag/D)	EE72_4.CIG	0.798	0.044	0.543	0.273	0.04
Silicon (Si/SiO$_2$)	ECLCLR6.LOF	0.391	0.451	0.440	0.357	0.85
Fluorine-doped Tin Oxide (SnO$_2$:F)	LOW-E_4.LOF	0.819	0.113	0.732	0.109	0.15

3.2 Detailed Models and Validation

In order to evaluate and compare models for coated glazing, we need first to have an absolute standard for the angle dependence of a range of different coating types. Ideally, we would either use a reliable theoretical treatment or measure the values directly using a highly accurate instrument. Classical optical theory, i.e. Fresnel's equations modified for thin-film multi-layers, (Heavens, 1960) is of course applicable to this situation, but obtaining a good structural description or model of the specimen is not automatic. Some prior knowledge of the coating structure and composition, however, together with some accurate radiometric and ellipsometric data, is usually sufficient to refine the model.

A detailed structural model was obtained for each coating in the test set with the cooperation of the manufacturers. Transmittance and reflectance data from 250nm-2500nm was then generated using Fresnel's equations extended to many layers including thin-film interference. A sample piece of each glazing in the test set was measured using both the ellipsometric and spectrophotometric techniques over the same wavelength range. The experimental and model-generated ellipsometric and radiometric data were fitted together, weighting both data types according to their standard deviations. Backside reflections were generally accounted for and included as fit parameters. Minimization of the combined error function was by adjusting each layer in the model until the best MSE was obtained. This was an expected refinement as the exact layer thickness and optical constants are in general uncertain.

The detailed structural models obtained in this way are used to generate reflectance and transmittance data over the wavelength range 250 - 2500 nm at incidence angles from 0° to 90° in 5° steps. The spectrally averaged properties are then calculated at each angle and normalized to the measured data as described. These generated properties are assumed to be the "true" properties at each angle. To justify this assumption, the detailed models are used to produce angle-dependent curves at a single wavelength of 670 nm that can be compared to the accurate laser-diode goniometer measurements. Some absolute deviation is apparent for two of the three coatings shown in Fig. 1. Unfortunately, the instrument used to make the measurements at 670 nm is no longer in service and so the generated curves were from different samples of the same product. The materials of the old and new coatings are still nominally the same, but the thickness of the layers may be slightly different. In any case, the normalized profiles are in excellent agreement for all coatings in the set for both transmittance and reflectance.
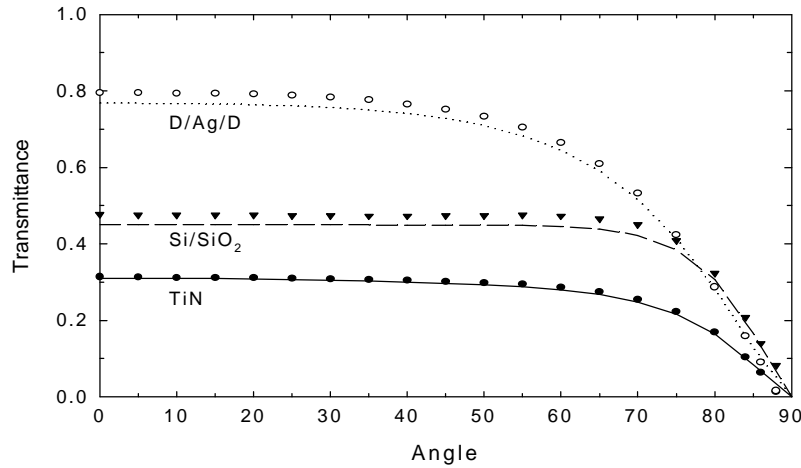

Fig. 1. Measured transmittance at 670nm using the laser-diode goniometer (symbols) compared to the data generated from detailed models (lines) of some materials in the test set.

3.3 Simplified Models

From the detailed models of the previous section, we can get a good idea of the range of possibilities that must be considered in developing simplified models. Uncoated glasses occupy a central band in the plots of Fig. 2. As the glasses become more absorbing, the transmittance curves fall, of course, and they also become flatter. The angular profile of coated glazing can fall significantly above or below this band; at 60°, the spread is about 40%. On the other hand, no practical coating is likely to deviate even farther from the uncoated band. We have tested filters with far more complex structures without finding additional variance. Only materials with some lateral microstructure will produce profiles that differ greatly in shape and magnitude from planar coatings. For example, angle-selective coatings with a louvered internal structure have been deposited (Smith, 1990; Le Ballac *et al.,* 1995).
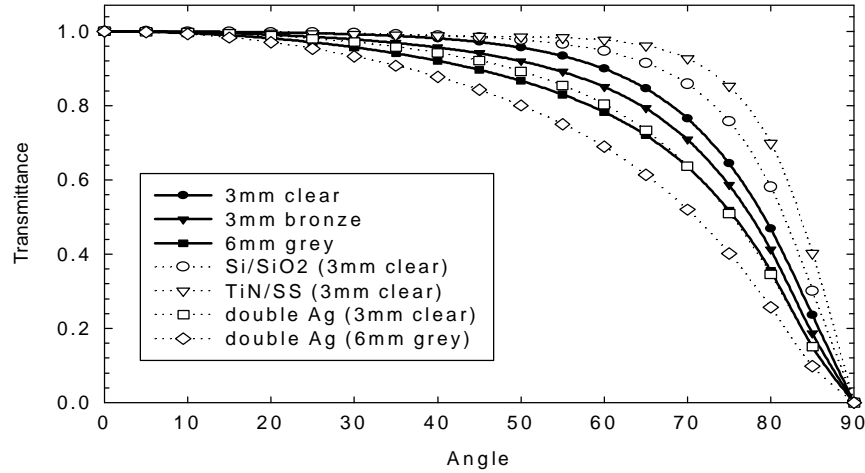
Fig. 2. Range of angular variation for monolithic materials and planar coatings.

Having established a database of accurate properties using the detailed models, it is now possible to evaluate the results of more practical models. Only the simplest structural models can be solved in closed form and these are often far from the actual structure of thin-film coatings. Realistic models for complex thin-film structures require a numerical solution similar to that of Section 3.2 but with further constraints. Unlike the unrestricted models discussed above, data will now be limited to the three radiometric values (transmittance, and front and back reflectance) at each wavelength that are found in each file of the Window 4.1 library.

3.3.1 Empirical and Semi-empirical Models

Many computer programs for window analysis in the public domain apply a purely empirical function, often a polynomial, to angle-dependent data. Without any physical basis, each set of empirical parameters would be expected to apply to only one material or to a class of very similar materials. Montecchi and Polato and Roos have proposed semi-empirical functions that have been accurately fit to experimental and generated data. This approach is appealing, because it reduces the representation of the angle-dependent properties to a few parameters. It has also been demonstrated that these semi-empirical models, having some basis in theory, apply more broadly than purely empirical functions. Physical models of coating structures would extend the flexibility of the approach even further. A broader range of materials and thickness should be possible. Also, a physical model would not necessarily be confined to a description of angle dependence. For example, knowledge of the effective optical indices permits the construction of more complex structures such as coated laminates.

3.3.2 Monolithic models

An uncoated glazing material such as a piece of thick glass can be modeled realistically as a single homogeneous layer without surface roughness or other inhomogeneity. In this case, there is an exact solution to the problem: Expressions for the measured values of R

6

and T can be written after accounting for multiple reflections and passes through the material. These expressions can be solved for the surface reflectivity and internal absorptance (Rubin *et al.,* 1988). Then Fresnel's law and Beer's law can be inverted to find the optical indices. From the calculated optical indices, all of the radiometric properties can then be determined as a function of wavelength, angle, and polarization. Sufficient accuracy for this procedure was confirmed experimentally by Petit (1979), (although he solved for the optical indices numerically, rather than utilizing the closed-form solution). There is no reason, then, not to use this model in all cases of monolithic glazing.

The simplest models that can be used for coated glazing apply the angular dependence of clear or tinted glass to all glazing materials. As shown in Fig. 2, this would not work particularly well even for other uncoated glasses of different tint (Furler, 1991). The monolithic model would not seem to be a much better description of the true structure and index distribution in a coated glazing. For spectrally averaged properties, however, the monolithic model gives results that are surprisingly good for some coated materials, as we will demonstrate.

There is more than one way, to apply the monolithic model to a coated glazing, so we will need to be more specific. Unlike a true monolithic material for which reflectance is the same from each side, a coated material is asymmetric. One possible solution then is to average the two values of reflectance, then apply the monolithic model, assigning the same angular profile to each side. A second approach is to use the reflectance from each side, perform the calculation twice, and apply a different angular profile to reflectance from each side. The latter approach might seem more logical, but both models are clearly far from reality, so we shall reserve judgment.

3.3.3 Thin-film models

In this section we describe a set of three very simple thin-film models with either one, two, or three layers. It was our hope that one or more of these models would converge for each coating type given the limited amount of commonly available optical data. Each of these three thin-film models is constrained to only two adjustable parameters. Although this stringent condition was not always necessary to obtain convergence (see Sec. 3.3.4), no operator intervention was required in any case. Less constrained models, possibly including a dispersion formula, gave better fits and more accurate results in most cases; the cost was a sequence of discrete fitting steps performed by an experienced analyst. If the sequence of steps can be programmed into some type of "expert system" then it might eventually be practical to use the more advanced models.

The simplest thin-film model describes the coating as a single layer of unknown thickness and optical indices. Even with this simple structure, we are required to make broad assumptions about either the material or its thickness. Best results were obtained with a fixed thickness of a metal. Both noble and transition metals as well as semiconductors are present in our test set. Fortunately, it was found that when this model converged it did so uniformly and regardless of the initial optical constants. The results that follow are based on starting values for the optical constants of silver (Lynch and Hunter, 1985) with a fixed thickness of 10 nm.

As the number of layers increases with the number of fixed parameters held constant, the models unfortunately become more specific. Assuming as usual that the substrate is known, two or three homogeneous coating layers are postulated. In the case of the double layer, the material and thickness of one of the layers must be specified. We chose to roughly base this model on TiN/SS, which has stainless steel and titanium nitride TiN layers (Fig. 3). The top layer was fixed as 10nm of TiN. The optical constants of the bottom layer are determined in the fitting procedure with the starting values set to those supplied for stainless steel. The thickness of the bottom layer is fixed as 10nm.

TiN	100Å
s.steel *	100Å
glass	

Fig. 3. Double-layer model based on transition-metal solar control coating.

In the case of the triple layer model, we chose a generic silver multilayer structure (Fig. 4) on a known substrate, similar to the structure of D/Ag+/D and D/Ag/D. The starting values were 20nm TiO_2/ 10nm Ag/ 20nm TiO_2. Only the optical constants of the middle layer were determined in the fitting procedure again starting with the values used for Ag in the single-layer model.

TiO_2	200Å
Ag *	100Å
TiO_2	200Å
glass	

Fig. 4. Triple-layer model based on a silver-dielectric low-emittance coating.

3.3.4 model convergence

The models described in the previous section were first independently fitted to the data associated each wavelength, then to data at all wavelengths simultaneously. Not all models converged to a solution for each glazing, which is an expected consequence of using such highly constrained models. The choice of starting values for the free parameters in each model can also influence the convergence. Montecchi *et al.* (1997) demonstrated that different fitting techniques could produce significantly different results for the optical properties.

The monolithic model always has a solution, so it can be used for all the glazing materials. Remember that, despite guaranteed convergence, this crude physical description may be highly inaccurate. The monolithic model is surely limited in at least one potentially important way because it cannot account for interference effects. Therefore the results at any given wavelength must be nonsense and only the spectrally averaged quantities have any chance at accuracy. Like the monolithic model, but without the limitation on interference, the one-layer thin-film numerical model also seems to be sufficiently general to allow convergence for each glazing in the test set. Again, convergence occurs despite the fact that some of the coatings are known to be considerably more complex than the model. Two-layer and three-layer models tend to converge to solutions only when applied to a glazing with sufficiently similar composition or structure.

An exception to this rule, the three-layer model converges for TiN/SS and SS, which are nominally 2-layer coatings. Possibly some inhomogeneity in one of the layers or surface roughness results in a structure more complex than is apparent. The double-silver stack, the most complicated coating in the test set, presents another apparent contradiction. Neither the 2-layer nor the 3-layer model converges for this coating despite similarity of

starting materials. Possibly, the models have become too specific for this complex structure whereas the relatively unconstrained single-layer model fits. Models that converged to a solution for each glazing were used to calculate spectrally averaged properties from 0° to 90° and then each profile was scaled to match the radiometric data at normal incidence.

4. Results and Discussion

4.1 Monolithic Model

It might seem that the asymmetric monolithic model would give better results than the symmetric monolithic model because coated glazing is, in fact, asymmetric, but this is not true in general. Results of the more complex models will then be compared only to the symmetric monolithic model. The extreme discrepancy between the assumed structure and the actual coating structure might be expected to produce highly inaccurate results. This expectation is not fully realized, as indicated by the results summarized in Table 2, but accuracy may not good enough across the board for many purposes.

Deviations of the monolithic model from the detailed model are given at 60° (Table 1). At this practical angle the projected angle factor $\cos\theta$ falls to 1/2 and the hemispherical weighting $\cos\theta\sin\theta$ is nearly a maximum at 0.43 . The deviations increase strongly at higher angles up to an angle of maximum deviation (Table 3), which lies somewhere between 75° and 85° depending on the coating. At 80°, however, $\cos\theta$ and $\cos\theta\sin\theta$ have both fallen below 0.2 and the large deviations are thus of reduced concern for energy calculations. Pfrommer, found deviations in heating and cooling loads up to 19% when using inaccurate angle dependent properties (Pfrommer *et al.,* 1994).

In this case, errors up to 5% and sometimes higher, as found at 60°, might seem quite acceptable for predictions of energy consumption and illumination. A desirable expectation is that the predictions will not deviate from the true value by more than 1%, which is typically achievable measurement accuracy at normal incidence. On this basis, the symmetric monolithic model fails to give acceptably accurate results for any glazing. This may seem to be an overly stringent criterion considering the larger uncertainties often encountered in simulations of building performance. Nevertheless, we use it as a target for now. Errors are often cumulative and there is no reason to incur them unnecessarily, when the goal seems achievable. Furthermore, manufacturers often use these properties for product comparison, and they would probably not be content with 5 percent error when they can get 1% at normal incidence.

In general the errors are greatest for the materials with the most complex structures such as the silver multi-layers, and least for the transition metal coatings such as TiN which has the simplest structure. For D/Ag/D/Ag/D, the deviations are always larger for the solar averaged properties. This type of coating is intended to be as close to the transparency of glass as possible in the visible, so that the monolithic model works fairly well in that region. The monolithic model holds true in the visible, to a lesser extent, for the other low-emittance coatings: D/Ag/D, SnO2:F and D/Ag+/D, which have a much less pronounced spectral transition. The opposite trend occurs for Si-based ECLCLR with the higher deviations occurring in the visible.

Table 2. Deviation of the prediction made by the symmetric monolithic model from the prediction made by the detailed model in percent. All values are at 60° angle of incidence.

Coating	Tsol	Rsol	R'sol	Tvis	Rvis	R'vis
SnO2:F	2.13	-0.90	-0.15	0.76	0.36	1.26
D/Ag/D	3.04	-4.98	-3.15	1.18	-3.34	-3.16
D/Ag/D/Ag/D	5.83	-7.84	-3.41	1.74	-3.31	-1.99
D/Ag+/D	1.68	-4.43	-3.74	-0.43	-3.85	-2.76
TiN	0.15	1.08	-1.59	0.28	1.12	-1.42
SS	0.68	-0.28	-2.00	1.29	-1.74	-2.47
TiN/SS	0.85	0.13	-3.07	0.75	-0.42	-3.10
Si/SiO2	1.50	-0.38	0.12	2.21	-1.07	-1.38

Table 3. Deviation of the prediction made by the symmetric monolithic model from the prediction made by the detailed model in percent. Values are at the angle of maximum deviation which falls between 75° and 80°.

Coating	Tsol	Rsol	R'sol	Tvis	Rvis	R'vis
SnO2:F	2.49	-1.57	0.80	1.30	2.39	3.48
D/Ag/D	8.24	-13.64	-9.91	3.17	-7.55	-6.98
D/Ag/D/Ag/D	15.72	-22.08	-13.10	4.80	-7.11	-3.97
D/Ag+/D	5.66	-12.75	-11.79	1.41	-10.12	-7.75
TiN	0.65	3.89	-8.84	0.98	2.49	-7.43
SS	1.90	0.37	-10.04	3.58	-5.39	-10.13
TiN/SS	2.59	1.10	-14.58	2.58	-1.82	-12.74
Si/SiO2	4.61	-1.43	-3.34	8.30	-2.84	-8.67

Some trends in this data are evident. The lower errors in the visible can be explained by considering that these coatings behave like different materials in the two regions of the spectrum. In the visible they are designed to have properties like clear glass, while in the solar infrared their properties become metallic. The monolithic model fares better in the visible part of the spectrum where the index distribution in the glazing is relatively smooth, but is inadequate in the solar infrared where there is a large difference between the substrate and coating index.

4.2 One-Layer Model

The single-layer numerical model does not give a large improvement in accuracy over the monolithic model. This is somewhat surprising considering that the assumed structure is considerably more realistic for any coating than the monolithic structure. Furthermore, the single-layer model is not highly constrained in the sense that the number of available data points is still greater than the number of adjustable parameters. Nevertheless, as Table 4 shows, the deviations for the single-layer model are rarely much smaller and in some cases larger than for the monolithic model. Based on our criterion of achieving accuracy similar to that measured at normal incidence, the errors are still unacceptably large for all coatings in the test set.

Table 4. Deviation of the prediction made by the one layer model at 60° from the prediction made by the detailed model in percent

Coating	Tsol	Rsol	R'sol	Tvis	Rvis	R'vis
SnO2:F	3.14	-0.26	1.01	-0.51	1.81	2.66
D/Ag/D	3.02	-4.54	-2.34	1.07	-2.70	-2.52
D/Ag/D/Ag/D	5.84	-7.67	-2.79	2.30	-2.85	-1.39
D/Ag+/D	3.65	-5.69	-2.18	3.61	-5.07	-1.98
TiN	1.93	-0.89	0.75	2.29	-0.40	0.80
SS	2.01	-2.23	0.67	2.56	-3.38	0.36
TiN/SS	1.38	-1.38	0.49	1.63	-1.87	0.18
Si/SiO2	1.69	0.00	1.70	0.61	0.18	1.50

Complex multi-layers are more poorly represented by the single-layer model than the simpler structures, as was the case with the monolithic model. Molina and Maestre (1996) saw a similar effect when analyzing coating similar to EE72.CRD and LOW-E.LOF. Even for TiN, which has only one coating layer, this model performs poorly. Since the coating thickness for TiN is far from the 10nm assumed in the one layer model, the optical constants obtained from the fitting procedure are far from their true values. The incorrect optical constants then give inaccurate predictions for the optical properties at oblique angles. For the other materials we must consider that the one layer model is still not close enough to the actual coating structure to give accurate predictions of angle dependent behavior.

4.3 Double-Layer Model

The double-layer transition-metal model only converges for coating types of similar composition and structure. As can be seen in Table 5, the deviations from the detailed model predictions are considerably improved over the previous two models. This reflects how much closer the model is to the actual structure of these coatings. The deviations at 60° fall within 1% of the detailed model for the two coatings (TiN and TiN/SS) that

contain TiN, which is the material set as the upper layer in the model. Constraining the model does make it specific to a class of coatings as expected, but it also significantly improves the accuracy of the angle dependent predictions to within our target values. Fig. 5 shows that the deviations for the simpler models are significant over a wide range of angle in the case of TiN. The monolithic model actually fits well in this case, but, from the progression of results so far, it is becoming clear that this is coincidental.

Table 5. Deviation of the prediction made by the double-layer model at 60° from the prediction made by the detailed model in percent

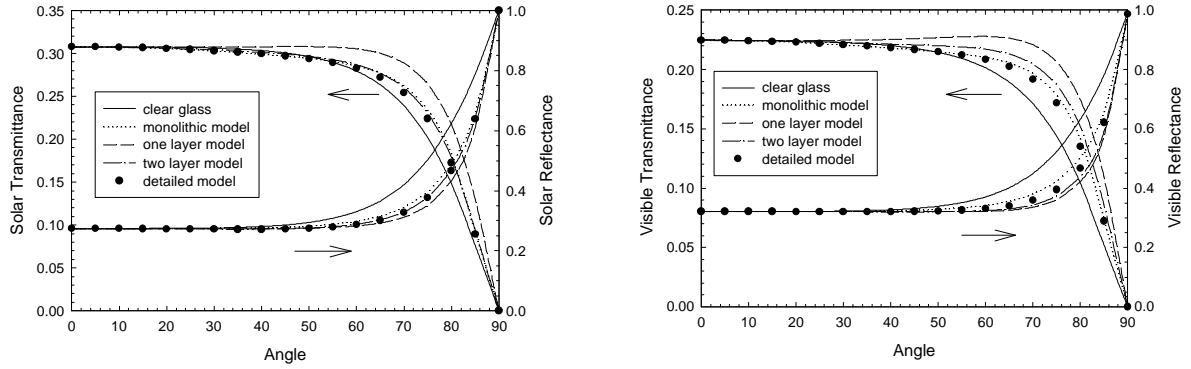|	Tsol	Rsol	R'sol	Tvis	Rvis	R'vis
TiN	0.85	-0.66	0.28	0.43	0.15	0.59
SS	0.76	-1.07	0.60	0.57	-1.39	0.71
TiN/SS	0.42	-0.78	0.02	-0.16	-0.28	0.32

Fig 5.. Angular properties of TiN compared to the 2-layer model and various other models.

4.4 Triple-Layer Model

The triple-layer model converges for the two coatings similar in structure and materials to the model, D/Ag+/D and D/Ag/D. This model also converges for TiN/SS and SS, which have simpler structures. The deviations from the detailed model predictions shown in

Table 5 are within the 1% limits for D/Ag/D, the glazing with the closest structure to the model. Although D/Ag+/D has a structure similar to D/Ag/D, the triple-layer model deviates further from the detailed model in this case. D/Ag+/D has a thicker silver layer than D/Ag/D, which affects the angle dependent properties significantly. Again, this contradicts previous observations that the thickness of the noble-metal layer has little effect on angular profile.

Table 5. Deviation of the prediction made by the three layer model at 60°
from the prediction made by the detailed model in percent.

Coating	Tsol	Rsol	R'sol	Tvis	Rvis	R'vis
D/Ag/D	-0.30	-0.50	-0.53	-0.64	-0.21	-0.31
D/Ag+/D	1.18	-1.60	-1.44	1.12	-1.81	-1.21
SS	0.62	0.18	-1.45	0.82	-0.15	-0.77
TiN/SS	0.16	0.85	-0.23	-0.05	1.34	-0.51

5. Conclusions

Detailed optical models were constructed and accurate angle dependent data was generated for a wide selection of coated glazing materials. This information can be used to confirm the validity of any proposed model of angle dependence. Among simplified physical models that can be used with standard data, only the monolithic model has an exact solution. As a realistic structural representation of uncoated glazing, the monolithic model is highly accurate, but far less so for coated glazing. Nevertheless, it presents a unified and fail-safe approach that is at least as good as currently accepted methods. A variety of very simple thin-film models were chosen for their expectation to converge. A single-layer numerical solution was not much better in general accuracy than the monolithic model and much harder to implement. At 60°, both the monolithic model and the single-layer thin-film model were often accurate to within 2% but frequently deviated farther up to 8%. Highly constrained multilayer models often deviated less than 1%, although they became increasingly specific to similar coating types. There are a variety of numerical techniques as well as the use of physical models of dispersion that can greatly improve these fits. The disadvantage of these advanced approaches is the increased probability that the process will not converge automatically. With techniques to ensure convergence, these results indicate that less constrained models could lead to a prediction of angle dependence with accuracy similar to that of measured properties at normal incidence.

6. Acknowledgements

7. References

ASTM E 903-82 (revised 1988) *Standard Test Method for solar Absorptance, Reflectance, and Transmittance of Materials using Integrating Spheres*, American Society for Testing and Materials.

Furler, R.A. (1991). Angular dependence of optical properties of homogeneous glasses. *ASHRAE Trans.* **97,** 1129-1133.

Heavens, O.S. (1960). Optical properties of Thin Films. *Reports on Progress in Physics XXIII.* **66**.

Le Bellac, D., Niklasson, G.A., and Granqvist, C.G. (1995). Angular-selective optical transmittance of highly transparent Al-oxide-based films made by oblique-angle sputtering. *J. Appl. Phys* **78,** 2894-2896.

Lynch, D.W. and Hunter, W.R. (1985). Comments on the optical constants of metals and a n introduction to the data for several metals. *Handbook of Optical Constants of Solids*, E.D. Palik, (Editor), Academic Press p. 350.

Molina, J.L. and Maestre, I.R. (1996).Spectral and Angular Characterization of Coated Glazing, *IEA Task 18 Report T18/A2A3/SP2/96*, International Energy Agency.

Montecchi, M. and Polato, P. (1995). Simple equations to foresee the daylighting behavior of glazing by normal incidence spectrophotometry. *Rivista della Stazione Sperimentale del Vetro.* **5,** 165-173.

Montecchi, M., Piegari, A., Peron, F. and Polato, P. (1994). Spectrophotometric characterization of homogeneous and inhomogeneous thin films for architectural glazing applications, *Glass Technol.* **35.**

Montecchi, M., van Nijnatten, P.A., Polato, P. and Roos, A. (1997). Spectrophotometric characterization of coated architectural glass sheets and its role in the simulation of daylight behavior of glazing. *Submitted to Glass Technology.*

NFRC 300-94: Procedures for Determining Solar Optical Properties of Simple Fenestration Products. *National Fenestration Rating Council.*

Optical Data Library for Use with the Window 4.1 Program, Addendum #5, *LBL Report 35298*.

Petit, R.B. (1979). Hemispherical transmittance properties of solar glazings as a function of averaging procedure and incident angle. *Solar Energy Materials*. **1,** 125-140.

Pfrommer, P., Lomas, K.J. and Kupke, Chr. (1994). Influence of transmission models for special glazing on the predicted performance of commercial buildings, *Energy and Buildings.* **21,** 101-110.

Pfrommer, P. , Lomas, K.J., Seale, C. and Kupke, Chr. (1995). The radiation transfer through coated and tinted glazing. *Solar Energy*. **54,** 287-299.

Roos, A. (1997a). Optical characterization of coated glazings at oblique angles of incidence: measurements versus model calculations, *J. Non-Crystalline Solids.* **218,** 247-255.

—— ——. (1997b). Optical Properties and Scattering: Final Project Report, *International Energy Agency Task 18*, *T18/B11/FPR/97* p. 17.

Rubin M., von Rottkay, K., Wen, S.J., Ozer, N. and Slack J. (1996). Optical Indices Of Lithiated Electrochromic Oxides. Paper presented at SPIE International Symposium on Optical Materials Technology for Energy Efficiency and Solar Energy Conversion XV, Freiburg, Germany. *To be published in Solar Energy Materials*.

Rubin, M., Powles, R. and von Rottkay, K. (1988). Window Optics. *Solar Energy*. **62.**

Ruzakowski Athey, P., Tabet, M.F., and Urban III, F.K. (1997). Determining the optical properties of a mixed-metal oxide film, Co3-x-yCrxFeyO4, with spectroscopic ellipsometry and atomic force microscopy. *Journal of Vacuum Science and Technology.* **A 15**(3), 998-1006.

Ruzakowski Athey, P., Urban III, F.K., Tabet, M.F., and McGahan, W.A. (1996). Optical properties of cobalt oxide films deposited by spray pyrolysis. *Journal of Vacuum Science and Technology.* **A 14**(3), 685-692.

Smith, G.B. (1990) Theory of angular selective transmittance in oblique columnar thin films containing metal and voids. *Applied Optics.* **29** 3685-3693.

von Rottkay, K. and Rubin, M. (1996). Optical Indices Of Pyrolytic Tin-Oxide Glass. *Mater. Res. Soc. Symp. Proc*. **426,** 449-454.

von Rottkay, K., Rubin, M., and Ozer, N. (1995). Optical Indices Of Tin-Doped Indium Oxide And Tungsten Oxide Electrochromic Coatings. *Mater. Res. Soc. Symp. Proc*., **403,** 551-556.